



BRNO UNIVERSITY OF TECHNOLOGY

VYSOKÉ UČENÍ TECHNICKÉ V BRNĚ

FACULTY OF ELECTRICAL ENGINEERING AND COMMUNICATION

FAKULTA ELEKTROTECHNIKY
A KOMUNIKAČNÍCH TECHNOLOGIÍ

DEPARTMENT OF FOREIGN LANGUAGES

ÚSTAV JAZYKŮ

NANOSTRUCTURES AND NANOSENSORS

NANOSTRUKTURY A NANOSENZORY

BACHELOR'S THESIS

BAKALÁŘSKÁ PRÁCE

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BRNO 2017

Bakalářská práce

bakalářský studijní obor **Angličtina v elektrotechnice a informatice**

Ústav jazyků

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ID: 173588

Ročník: 3

Akademický rok: 2016/17

NÁZEV TÉMATU:

Nanostruktury a nanosenzory

POKYNY PRO VYPRACOVÁNÍ:

Charakterizujte nanostruktury (jejich dělení, vlastnosti a výrobu) a popište jejich použití ve snímacích zařízeních. Analyzujte využití nanosenzorů v praxi.

DOPORUČENÁ LITERATURA:

- 1) Natelson, D. (2015). Nanostructures and nanotechnology. Cambridge: Cambridge University Press.
- 2) Bhushan, B. (2012). Encyclopedia of nanotechnology. Dordrecht: Springer.
- 3) Fojtík, A. (2009). Nanostruktury a nanotechnologie - jejich aplikace a možnosti. Nanostruktury - důmyslné formy hmoty pro nové technologie. Praha: České vysoké učení technické.

Termín zadání: 6. 2. 2017

Termín odevzdání: 2.6.2017

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Abstract

The aim of this bachelor's thesis is to introduce the application of nanotechnology in sensing devices, provide fundamental information about nanotechnology including the description and the classification of nanostructures, and emphasise the possibility to enhance sensor operation by means of nanotechnology. The thesis also deals with the application of a nanowire as part of a sensor using the configuration of the field-effect transistor. Quantum dots and their application in optical detection, mainly in the field of nanomedicine, are considered in this thesis as well. The last part is focused on the advantages and current restriction of nanosensors.

Key words: nanotechnology, nanosensor, nanowires, quantum dots, detection

Abstrakt

Cílem této bakalářské práce je představit využití nanotechnologie ve snímacích zařízeních, poskytnout základní informace o nanotechnologii, včetně popisu a klasifikace nanostruktur, a zdůraznit možnost vylepšení výkonu senzoru prostřednictvím nanotechnologie. Tato práce se také zabývá využitím nanodráty jako součástí senzoru s konfigurací unipolárního tranzistoru. Kvantové tečky a jejich využití v optické detekci jsou také zahrnuty v této práci. Výhody i současná omezení nanosenzorů jsou shrnuty v závěru práce.

Klíčová slova: nanotechnologie, nanosenzor, nanodráty, kvantové tečky, detekce

KŘEČKOVÁ, J. *Nanostructures and nanosensors*. Brno: Vysoké učení technické v Brně, Fakulta elektrotechniky a komunikačních technologií, 2017. 41 s. Vedoucí bakalářské práce Mgr. Ing. Eva Ellederová.

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V Brně dne

.....

(podpis autora)

Poděkování

Děkuji vedoucí bakalářské práce Mgr. Ing. Evě Ellederové a odbornému konzultantovi doc. Ing. Pavlu Šteffanovi, Ph.D. za účinnou metodickou, pedagogickou a odbornou pomoc a další cenné rady při zpracování mé bakalářské práce.

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Introduction and aims

This bachelor's thesis investigates nanosensors based on various nanostructures as one of the emerging technology at the present time. Recently, the research of nanostructures has been of great importance promising many breakthroughs that may offer the improvement in several applications, new technological opportunities as well as new challenges.

Nanotechnology is an interdisciplinary field, which studies the particles with size less than 100 nm providing nanostructures with novel properties and behaviour. Therefore, the main focus in this study is the description of some types of the nanostructures, including the process of fabrication and characteristic properties, together with the aim to introduce its application in practice as a nanosensor.

The Bachelor's thesis is divided into five main chapters, where the first one attempts to summarize some basic definitions and properties connected with the nanostructures that should be mentioned and discussed in order to provide better comprehension of this topic. The second chapter deals with the introduction to sensor's technology, parameters of a sensor and its parts, in particular, are described.

After being acquainted with the basic principles of both nanotechnology and sensor's technology, we can proceed to a combination of these two phenomes, i.e. nanosensors. Therefore, the third and the fourth chapters are dedicated to the description of nanosensors, where the nanowires and the quantum dots, respectively, will be discussed as representatives of nanostructures employed in sensor's technology.

When examining the possibility of the application of both the nanowires and the quantum dots in sensing mechanisms, the fabrication processes and properties of both nanostructures will firstly be described. Then, the latter parts of chapters 3 and 4 include the demonstration of the practical applications of the above-mentioned nanostructures in sensor's technology, including the description of working principle of such nanosensors.

The last chapter is focused on the advantages of nanotechnology-enabled sensors over common sensors, and provides examples of sectors in industry and services that will benefit from this modern technology. However, also restrictions that limit implementation of this technology into the industrial mass-scale production are listed and discussed there.

1 Introduction to nanostructures

As described by Fojtík (2008), nanotechnology and nanoscience deals with very small objects with the dimension of a nanometers scale and studies the properties this kind of matter exhibits. A study of such structures, called nanostructures, is a part of an interdisciplinary field of research where chemistry, physics, biology and mathematics overlaps in order to provide further information about methods of preparation and manipulation with such objects. Although, the term “nanotechnology” has been coined quite recently, the existence of nanostructures on the Earth is as long as life itself. For example, Poole (2003) mentions ancient glassmakers who used metals nanoparticles to fabricate glass providing great varieties of beautiful colours as the different size and shape of gold nanoparticles exhibit different colours.

1.1 Definitions and classification

In the *Encyclopedia of nanotechnology*, nanostructures are defined as “portions of a material where at least one spatial dimension is at the nanoscale” (Bhushan, 2012, p.1942). Another definition is provided by Fojtík (2008), where the fact that nanostructures also exhibit unique properties which do not occur in macroscale is emphasized. Nanoparticles, nanowires, nanotubes, and thin films are examples of nanostructure, all of them featuring novel properties including electric, optical, chemical, and mechanical ones. The dissimilarity of mentioned properties between nanoscale and macroscale structures is mainly based on the effect of quantum confinement of electrons in one or more spatial dimensions (Bhushan, 2012). The parameter called “surface-to-volume-ratio” is strongly influenced by miniaturization, thus also altering chemical characteristics. Those two phenomena will be further described in the following subchapters.

First of all, the classification according the space restriction will be mentioned, though. In nanospace, nanostructures can be restricted to one, two or three dimensions, which generally leads to changes in physical properties in the given direction. Depending on the number of dimensions outside of the nanometre range, or according to the

number of dimension in which electron motion is permitted, nanostructures are divided into zero-, one-, two-, three- dimensional structures (Kelsall, 2005). Examples of some nanostructures are illustrated in Figure 1.

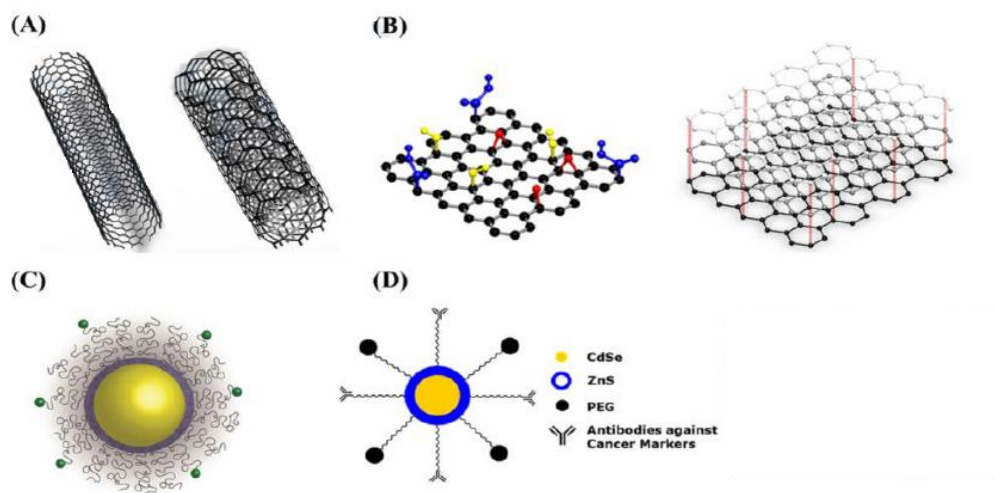


Figure 1. Illustration of some nanostructures (A) 1D – carbon nanotube (left – single-walled; right – multi-walled), (B) 2D – graphene (left – single-layered; right – multi-layered), (C) 0D – nanoparticle, (D) 0D – quantum dots.

Reprinted from Islam (2014, p. 48).

Structures that are confined in all three directions are called **zero-dimensional** as they do not permit electron motion in any direction. All three spatial dimensions are at the nanoscale, i.e. between 1 and 100 nm. This category includes nanoparticles and quantum dots.

One-dimensional structures are restricted to two dimensions leading to the electronic motion in one direction. Nanotubes and nanowires are examples of this class, featuring a characteristic diameter of nanoscale dimension and a length that could be of greater size. Alternatively, it can be called with the term “quantum wire” (Fojtík, 2008).

Analogously, **two-dimensional** structures are limited to one dimension enabling the movement of electrons in two directions. A representative example of this class is a thin film, which is two-dimensional with the thickness of tens of nanometers. A planar quantum well also belongs to this class.

Finally, **three-dimensional** structures are represented by bulky materials with dimensions above 100 nm, however, containing 0D, 1D and/or 2D structures. These structures include nanocomposites and nanostructured bulky materials, which are classified as crystalline materials, polycrystalline materials, and amorphous materials (Bhushan, 2012).

1.2 Effects of the nanometer length scale

As previously mentioned, the characteristics of nanoscale materials are markedly different from those in macroscale due to spatial confinement in one, two, or three directions. Kelsall (2005) notes that not only is the energy band structure influenced by this effect, but also atomic structures can be indirectly affected.

The energies of the electronic states depend on the dimension of the system, thus when the length reduction in any direction occurs, it leads to reductions of space for free electrons movement. Hence, the respective wavelength of electrons has to be shorten, which is the only way for the electrons to continue their existence as a part of such a structure. By shortening the wavelength, electrons increase their energy and thus generates a higher energy level in the system. Furthermore, as the system size decreases, the energy bands become narrower and electrons tend to behave more like the “particles in a box”, also called the phenomenon of quantum confinement, which is described by Fojtík (2008, 2014). As a consequence of a higher energy level, the physical properties, such as the solubility, colour and absorption of the system, begin to change. Another effect of this behaviour is a change of the total energy and hence the thermodynamic stability of the system that can result in implementing a different crystal structure from that of the bulk material.

In order to understand the changes in the nanoscale system, the proportion of atoms in contact with a free surface shall be taken into account. The surface area to volume ratio is the number of atoms on the surface in relation to the number of atoms in volume of an object. It is inversely proportional to particle size and thus increases drastically with reduced dimensions of the system (Kelsall, 2005). As the surface area per volume

of material increases, a greater number of atoms are on the surface and can come into contact with the surrounding materials, thus affecting reactivity.

A significant number of properties are size-dependent and hence affected by the quantum size confinement effect and also by the increase of the surface-to-volume ratio. When describing some of the properties, I will proceed from the *Encyclopedia of nanotechnology* (Bhushan, 2012) and from the work by Kelsall (2005), otherwise the reference will be stated.

As far as the influence of the quantum confinement is concerned, it is important to mention electronic properties. As a result of decreasing the system size below the critical length scale, the electronic states are formed in discrete energy values and the spacing between discrete states widens as the quantum confinement increases, i.e. size decreases. The details of discretization depend on the number of spatial directions that have been confined, therefore a fully discrete density of states is only observed in zero-dimensional. Therefore, “the average energy will not be determined so much by the chemical nature of the atoms, but mainly by the dimension of the particle” (Pool, 2003, p. 82). In certain cases, transition from conducting material to insulator can occur as the energy bands cease to overlap.

A change of the electronic structure due to the size reduction significantly influences the energies of the highest occupied molecular orbital (valence band) and the lowest unoccupied molecular orbital (conduction band), therefore altering optical emission and absorption, as these processes depend on transitions of electrons between these energy states. As a result, metals and semiconductors show large change in colour, for example colloidal solutions of gold nanoparticles have a deep red colour.

The large surface area to volume ratio has a profound effect on chemical properties as a chemical reactivity. Nanostructures are known to potentially adopt different crystallographic structures, which can be accompanied by an extraordinary increase in catalytic activity, for example in the case of a gold nanoparticle. Moreover, as the catalytic chemical reactions occur at material surface, a given nanomaterial will be much more reactive than its larger counterpart, in particular due to the significant surface area to volume ratio. This characteristic can increase the efficiency of catalysis

while reducing waste and pollution. Additionally, it has been found that some materials, originally inert in a macroscale, are reactive when produced in a nanoscale, consequently they are expected to interact with different kinds of substances enabling many potential applications including sensing technology.

Other important size-dependent properties important for sensor operation will be subsequently described in the chapters concerning relevant nanostructures.

2 Sensor Fundamentals

This chapter will provide the most fundamental information about sensors together with some important parameters for characterizing their performance.

To start with the definition of sensor itself, Wilson (2005, p. 1) defines it as “a device that converts a physical phenomenon into an electrical signal”. However, there is a difference between a sensor and a transducer, even though, in practice, these terms can be interchanged very often. With respect to differentiation of these terms, Wilson (2005, p.15) afterwards provides a more detailed definition: “Strictly speaking, a sensor is a device that receives a signal or stimulus and responds with an electrical signal, while a transducer is a converter of one type of energy into another.”

A transducer is, therefore, the sensitive part of the sensor that detects the analyte¹ and usually converts one type of energy into another. While sensors are systems that comprising of several components that combine tasks important for sensing, including a transducer, an amplifier, an A/D converter, along with information storage and processing. However, it depends on a type of the sensor and its complexity.

2.1 Parameters

The design of each type of sensor is dependent on the requirements placed on its application and use. In this subchapter, some of the most important parameters for characterizing sensor performance and their definitions will be introduced.

A suitable relationship of an input and output signal, represented as a graph, is in many cases the deciding sensor requirement. As Wilson (2005) states, the details of this relationship, called a transfer function, may fully describe the sensor characteristics. General demand on a sensor is high sensitivity, which is defined as the extent of change in a sensor's output when the measured quantity varies (Wilson, 2005). Another important parameter called dynamic range represents the range between the minimum

¹ Analyte is a substance (e.g. chemicals) that is being sensed (Evans, 2009).

and the maximum value that can be detected. Wilson (2005) notes that very inaccurate values occur when measured outside of this range. The ability of a sensor to interact only with the target analyte and reject the interferences of other substances is called selectivity. This feature is especially desired in the case of biosensors. And finally, as far as the resolution of sensors is concerned, it is defined as “the smallest reliable measurement that a system can make” (Wilson, 2005 p. 202).

2.2 Nanosensors

Sensor technology, as well as the other technological fields, is currently under the influence of the developments in nanotechnology, which promises a new enhanced design of sensors. According to Lim (2011, p. 7), “Nanosensors are sensing devices with at least one of their sensing dimension being no greater than 100 nm.” However, besides nanosensors we should also consider a “nanotechnology-enabled sensor”, which is a device that is enhanced by means of nanomaterials and is, for instance, able to detect the analyte of a nanoscale dimension. There is no doubt that the employment of nanotechnology offers advantages in terms of greater sensitivity and selectivity, as well as a smaller size connected with lower weight and lesser power requirements.

As a matter of course, nanosensors are promising devices for future sensor technology, possibly enabling to meet some objectives of sensor research and development. Evans (2009) emphasizes the following aims: targeted transducers, multiplexing, and multi-parameter transducers. The call for targeted transducers lies in the problem of a complex environment containing diverse types of molecules. As Evans (2009) proposes, nanotechnology has the potential to enable targeted transducers to selectively target and measure the specific analyte over different levels of concentration in multi-element environment.

Moreover, nanotechnology would bring the ability to simultaneously detect several analytes, for example toxic chemicals. This ability is called multiplexing and as Evans (2009 p. 4) explains, “A sensor could contain multiple nanoparticle species, each able to target a specific molecule.” Finally, multi-parameter transducers would provide

a measurement of several parameters, i.e. physical, chemical, or biological properties, thus deliver better differentiation of the target analyte.

The application of nanostructures and their novel properties is particularly favourable as far as biosensors are concerned. The biosensors, used for molecules detection and monitoring, consist of “transduction of some desired chemical or physical effect (e.g., the presence of a cancer marker protein) into some readily detectable signal (e.g., a change in electrical conduction, a change in optical properties)” (Natelson, 2015, p. 534). Besides the ability to detect a signal, great importance is placed on the specificity of the sensor, in other words, the ability to recognize the source of the signal. Biosensors are very useful tool for diagnosing in medical treatment, both for *in vivo*² and for *in vitro*³ sensing.

Natelson (2015) above all highlights two examples of biosensors with employed nanotechnology. The first one, and according to Natelson (2015) the most used, is a nanoelectronics sensing approach. It is based on the changes of conductance due to a change of chemical property. Possible implementation is via FET (field effect transistor) layout, using a semiconductor nanowire or a carbon nanotube as a conductive channel. This configuration will be discussed in the following chapter (3 Nanowire-based sensors).

The second one is a nanophotonic sensing approach, which takes advantage from unique optical properties of quantum dots (0D nanostructure) together with using a FRET (Förster resonant energy transfer) phenomenon. This phenomenon, as well as quantum dots, will be further described in the fourth chapter (Quantum dots), though.

² *In vivo* means in living multicellular organism (Natelson, 2015).

³ *In vitro* means in prepared samples of biological material (Natelson, 2015).

3 Nanowire-based sensors

As already mentioned, nanostructures exhibit novel properties due to their small size, therefore, the first part of this chapter is dedicated to a description of such parameters along with the fabrication process of a particular nanostructure, in this case of a nanowire, abbreviated as NW. The second part of this chapter deals with the use of the nanowire in sensing applications and the explanation of each type of employment.

3.1 Properties and fabrication of nanowires

3.1.1 Properties

There are many types of NWs available and they can have different properties depending on the material used for the fabrication. Islam (2014) notes that nanowires can be metallic (e.g. Pt), semiconducting (e.g. Si), and insulating (e.g. SiO₂), and therefore find possible applications in various fields.

In the first chapter, we have defined the classification of nanostructures according to the number of dimensions outside of the nanometre scale. Nanowires have the diameter at the nanometer scale and the length at macroscale, which makes them example of one-dimensional category. As nanowires possess two dimension in nanoscale, electrons and holes motion experience quantum confinement effect, but still one dimension, the length, is unconfined, thus allowing electrical conduction in metallic and semiconducting nanowires. Besides, nanowires have an extraordinary aspect ratio, i.e. length-to-width ratio, as a NW has the diameter of just a few nanometers and its length reaches up to hundreds of micrometers (e.g. diameter of human hair), which is the length visible even for the naked eye.

The high aspect ratio is also accompanied by high surface-to-volume ratio, which is by far the most promising quality for sensors application. This is also supported by Chandler (MIT News, 2013) who explains that the whole surface can be used to detect the specific molecule. The detection would create a signal, whose transmission along

the wire would be performed. Moreover, Chandler (MIT News, 2013) considers the macroscopic length of NW as a possibility of connecting a nanoscale device with a macroscopic one.

3.1.2 Fabrication

The fabrication of nanostructures in general can be divided into two approaches: a bottom-up and top-down approach. While in the bottom-up approach, atoms and molecules are manipulated as a building block to create a larger structure, the top-down approach uses a contrary technique that is based on cutting materials into the desired shape with the use of externally controlled tools. Both approaches include several diverse methods, but when considering synthesis of nanowires, the vapor-liquid-solid (VLS) method and the templating method are commonly employed.

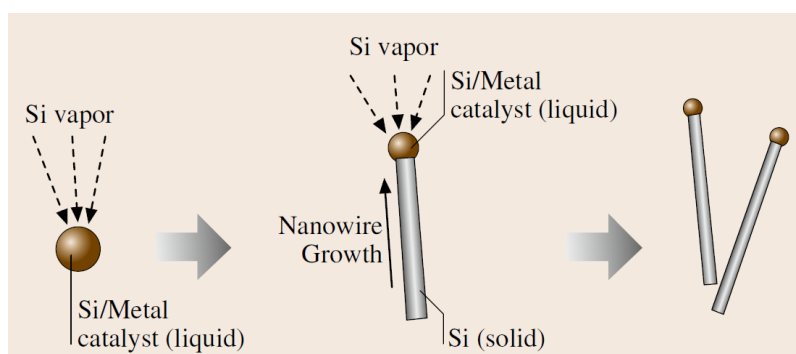


Figure 2. Illustration of Si NW growth by the VLS method.

Reprinted from Bhushan (2010, p. 125).

The VLS method, as an example of the bottom-up approach, is widely used to fabricate semiconductor nanowires and is further explained by Bhushan (2010) as well as by Dan et al. (2008). A schematic illustration of VLS method can be seen in Figure 2. The mechanism is based on thermal evaporation, when source material is heated until the evaporation process is reached. Then, the vapour is brought to the substrate where with the help of catalysts the nanowire growth occurs. Both authors present a gold particle as a metal catalyst, and an oxide, or a sulphide alternatively, as a non-metal catalyst. Either type of catalyst reacts with vapour and forms a liquid droplet on the substrate. This place on the substrate becomes “the nucleation site” once the droplet is supersaturated, i.e. containing more dissolved solute than is normally possible under

given conditions of temperature and pressure (thefreedictionary.com). The nucleation sites, as Bhushan (2010) also explains, are places where solid nanowires are generated through precipitating the source material. Dan et al. (2008) further adds that as long as the catalyst, located on the tip of the growing NW, remains liquid, the process of growing continues, thus the length of nanowire can be controlled.

To briefly describe the second method for nanowire fabrication, the templating method, I will also proceed from the description provided by both Bhushan (2010) and Dan et al. (2008). The templating method is based, as the name implies, on the utilization of the template with nanopores of a cylindrical shape. The desired material, which the NW will be synthesized from, is then by means of electrochemistry used to fill the empty spaces. The material then copies the shape, and in the end, in order to obtain formed nanowires, the template is dissolved. Both authors introduce an anodic aluminium oxide membrane as the most applicable template for metallic and semiconductor nanowires.

3.2 Nanowires in sensing applications

As a matter of fact, sensors find various applications in our lives and one of such applications is in medicine – detection of diseases and diverse chemical molecules. Yet, commonly used sensors are not sensitive enough to detect small concentration of molecules. Several researches in the field of sensor technology with the connection to nanotechnology have been done so far and as a result, many enhancements are available today. Bhushan (2010, p.157) expects sensor to “be smaller, more sensitive, demand less power, and react faster” with the help of nanotechnology. In this section, the application of semiconductor and metal-oxide nanowires as field-effect sensors will be described.

3.2.1 Nanowire field-effect sensors

The principle of the nanowires in application as a field-effect sensor is clearly described by Chen (2011), however, only silicon nanowires are considered. As Chen (2011) explains, a detection when using the field-effect transistor (FET) consists of three

electrodes, including source, and drain electrodes, which are connected through a conductor channel, and the third one, a gate electrode regulating the conductance of the channel. Semiconductor nanowires, made from silicon, serve as a conductor channel so as the sensing mechanism is possible. The schematic drawing of the nanowire FET sensor is shown in Figure 3, where either the nanowire as an individual channel (a) or the network of nanowires (b) serves for active detection.

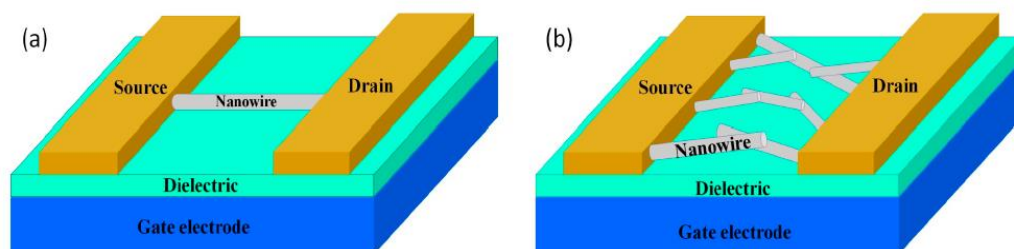


Figure 3. Schematic drawing of the nanowire FET sensor.

Reprinted from Feng (2014, p.4).

Feng (2014) further clarifies the operation of sensors that is based on the change in the FET parameters upon detecting the target analyte. Semiconductors, in general, are of two types depending on the charge carriers: n-type (electrons) and p-type (holes). Therefore, the alteration of FET parameters is also dependent on the type of semiconductor, which Chen (2011) thoroughly describes and depicts in his review. Considering the p-type channel, when the analyte with a positive charge is detected, the conductivity decreases because of depletion of carriers in the nanowire. Analogously, the detection of the positively charged analyte results in increase of the conductivity and vice versa in the case of the n-type channel. Often, further surface modification of the nanowires is required in order to enhance reactivity. Surface coatings, or a nanowire with an attached antibody are examples of such modification. Then, not only is the direction of change (increase/decrease) characterized, but also the interaction between the antibody and the analyte determines the magnitude of the change, as Chen (2011) adds.

A research of biomolecules detection using nanowires which were modified by an attached antibody was reported by Chen (2011) and Bhushan (2010). In this research,

the interaction of protein streptavidin with biotin-modified Si NW was studied. Thanks to the modification of Si nanowires with biotin, the FET sensor could detect the protein at very low concentration. Furthermore, the p-type Si nanowire FET sensor was investigated for the detection of DNA, where PNA (peptide nucleic acid) is attached to Si-NW. Chen (2011, p. 144) additionally explains: “PNA, an artificially synthesized polymer similar to DNA, is commonly used in biological research, especially in DNA or RNA hybridizations, i.e. PNA hybridizes with DNA by base pairing through hydrogen bonds.” Since DNA and RNA are negatively charged molecules, the hybridization with PNA results in conductance increase. Chen (2011) also highlighted the ability for microRNAs detection, since several cancers and neurological diseases are associated with deregulation of this molecule. In this investigation, the Si nanowire FET sensor with PNA modification is introduced as a promising tool for medical diagnostic, including early cancer detection or other disorders.

Besides the above-mentioned applications, nanowire FET sensors are also used for gas detection using n-type metal-oxide nanowires (SnO_2 , ZnO). The principle of sensing, as explained by Feng (2014), is rather the same as in the previous case, i.e. the interaction between the gas molecule and the channel will result in change of the FET parameters. One of the modified parameters is the carrier concentration, which is dependent on the surrounding environment and reacts differently in oxidizing and reducing gases. Feng (2014) reports that the oxidizing gas environment results in immobilization of free electrons thus leading to conductivity decrease, and vice versa for the reducing gas environment. Together with this modification, the concentration of carriers can be further tuned by the gate voltage thus improving the sensitivity. As far as the practical application is concerned, Feng (2014) introduces that further modification of the SnO_2 nanowire, in particular with a coat of palladium nanoparticles, makes the channel extraordinarily sensitive to hydrogen (H_2), a flammable and explosive gas.

In addition, Patolsky (2006) presents further applications, mainly in medicine. In his review, he considers the use of a nanowire-based sensor as a tool for drug discovery, since the specific binding of organic molecules to proteins is the key for the discovery of new pharmaceuticals. Patolsky (2006) also mentions the ability of nanosensors to detect a single virus with the help of an attached specific antibody to the nanowire. Studies confirmed that even a single virus causes a change in conductance when it binds

to the receptor – antibody, thus making the electrical detection of a single-particle possible. An extremely advanced application consists in integrating nanowire sensors into arrays, where diverse sensor devices utilize the modification of attaching specific antibody receptors, which binds with different viruses. In that way, as Patolsky (2006) suggests, selective multiplexed detection will be possible with a promising application in medical diagnostic as well as in defence.

4 Quantum dots

The following chapter investigates quantum dots⁴ as a promising nanostructure in sensing technology, more precisely in the field of nanodiagnostics as a part of nanomedicine. Nanotechnology-enabled diagnostics plays an important role in nanomedicines where it provides faster detection of pathogenic processes thus helping with the fight against many serious diseases. Quantum dots are particularly attractive when the optical detection is taken into account since photoluminescence is one of the most important optical properties of quantum dots. Moreover, the possibility to influence the light absorption and emission characteristics together with other unique properties make them suitable for substituting the organic dyes, which are currently used in biosensing.

Successively, the definition and main properties of quantum dots as well as approaches to the synthesis will be provided in the following subchapters, concluding with the final subchapter where the possible application of quantum dots as a tool for nanodiagnostics will be described.

4.1 Definition and properties

Quantum dots (hereinafter referred to as QDs) are nanometer-sized crystals composed of up to thousands of atoms of semiconducting material, yet only up to a hundred of free electrons (Fojtík, 2014). According to the classification of nanostructures that is included in the chapter 1.1 of this thesis, the QDs represent zero-dimensional (0D) structure with the typical size range between 2–20 nm (Drbohlavová et al., 2009), however, other authors state the range of the size below 10 nm, for instance Fojtík (2014) and Honeychurch (2014).

Nevertheless, the dimension itself is not efficient for determining whether the system is a quantum dot or not. Drbohlavová (2015) explains that the system can be determined as a quantum dot on the basis of the comparison of the nanoparticle radius and Bohr

⁴ Quantum dots are often abbreviated as QDs.

radius of electron (a_e)⁵. If the radius of a nanoparticle is lower than Bohr radius of electron, the effect of quantum confinement occurs and thus the system is called a quantum dot. However, the Bohr radius varies according to the used material, which confirms that, not the size itself, but also the material is a crucial factor when referring to QDs. The quantum confinement effect was, in general, described in the previous chapter (1.2) of this thesis, yet it is worth mentioning the properties which stem from this effect. Honeychurch (2014) points out that as a result of the confinement of charge carriers in all three dimensions, as in the case of QDs, the energy states and consequently the bandgap energies are size-dependent, i.e. with the increasing size the bandgap energy decreases.

As far as a structure of QDs is concerned, Hulicius (2012) emphasises the importance of distinguishing between nanocrystals made from single material, e.g. a gold nanoparticle, and QDs made from two different materials.

Even though QDs composed only of a semiconducting core exist, such as cadmium selenide (CdSe) and cadmium sulphide (CdS), they are usually not used in practice. As Drbohlavová et al. (2009) note, cadmium is a toxic element and it was reported as the cause of cytotoxicity. Therefore, in the core/shell organization, the toxic core is enclosed in a nontoxic shell made of different semiconductors in combinations like CdS/ZnS, CdSe/ZnS (Honeychurch, 2014), where the zinc sulphide (ZnS) represents material used for the shell as shown in Figure 4.

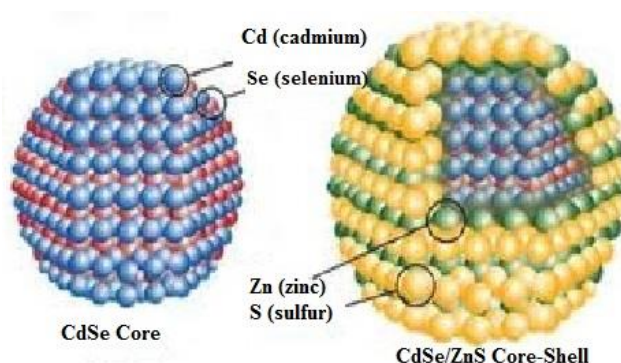


Figure 4. Diagram of CdSe core (left) and core-shell structure of CdSe/ZnS quantum dot (right). Reprinted from PortiaS, 2010.

⁵ Bohr radius is a distance between the centre of a nuclide and the electron in an atom at its ground state (lowest-energy level) (Wikipedia).

In this case, the shell can serve to reduce the toxicity of the core, it can make the binding of ligands⁶ possible, or simply enhance the optical properties, which will be furthermore described in the following subchapters.

4.1.1 Optical properties

As already mentioned in the introductory part of this chapter, the QDs have attracted significant attention mainly thanks to their unique optical properties. When a QD is radiated by UV or visible light, the excitation of electrons occurs, i.e. the electron is excited into a conduction band leaving a hole in the valence band creating an electron-hole pair termed an *exciton*. After a certain amount of time, the recombination of the electron with a hole appears upon which the electron emits energy in the form of light (Honeychurch, 2014), which is called *fluorescence*. In general, when comparing QDs with commonly used organic dyes, for instance fluorophore, QDs have “broad absorption spectra, very narrow emission spectra, long fluorescence lifetime, and high stability against photobleaching”, as Drbohlavová et al. (2013, p.554) summarize.

A prominent property caused by a quantum confinement effect is the tuneable and size-dependent light-emission as a consequence of the dependence of the bandgap energy on size. Since the bandgap energy equals to the energy emitted during electron-hole recombination, also the light emission colour varies as a function of size (Narlikar, 2010). Therefore, not only the material used but also the size of the core determines the light emission colour as illustrated in Figures 5 and 6 where the dependence on the chemical composition, as well as on the size, is shown, respectively. Moreover, the bottom part of Figure 6 shows the relevant size of a particle with diameters from 2.1 nm to 7.5 nm, from left to right (Torchynska, 2011).

By adjusting these two parameters, the light emitted by the QD can be tuned ranging from “the ultraviolet, throughout the visible, and into the infrared region (400–4000 nm)” (Narlikar, 2010, p. 616). The possibility to tune these properties allows us to select that emission wavelength which is suitable for the given experimental conditions.

⁶ Ligand is a molecule that binds to another (usually larger) molecule.

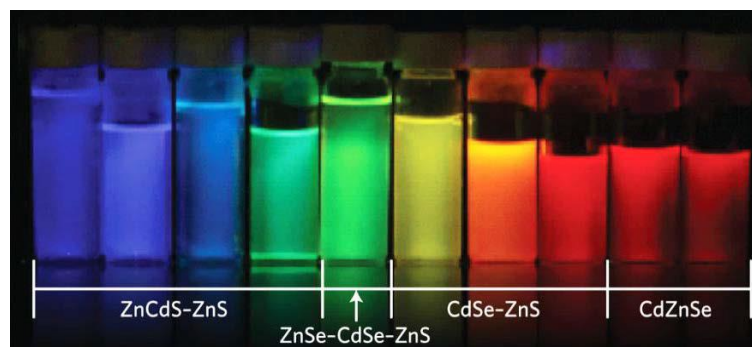


Figure 5. Dependence of the fluorescence spectra on the chemical composition of the QD core. Reprinted from Drbohlavová (2015, p. 13).

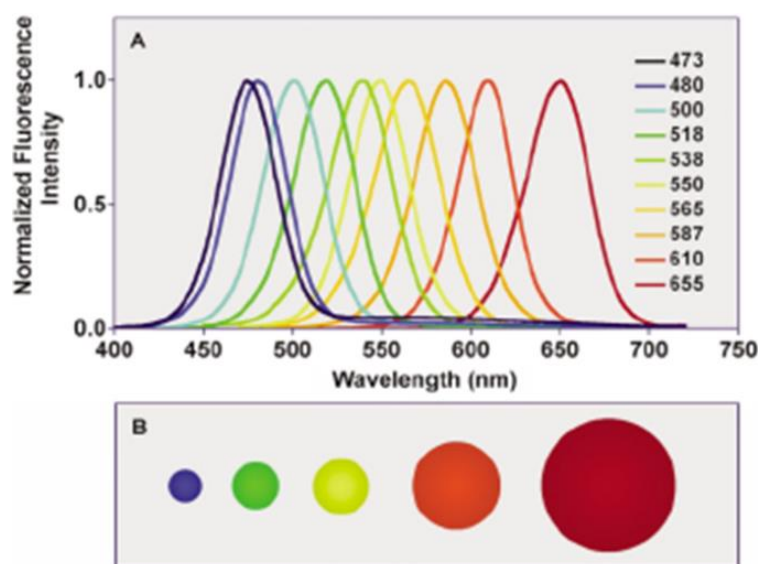


Figure 6. Dependence of the light emission colour on the size of the CdSe quantum dot. Reprinted from Torchynska (2011).

The unique optical properties of QDs are complemented with broader absorption spectra, i.e. they are more efficient absorbers. It is reported by Narlikar (2010) that QDs absorb light 10–50x faster than organic dyes resulting in higher brightness of QDs, which enables more sensitive detection. Drbohlavová et al. (2009) also add the possibility of excitation of various QDs by just one light source, e.g. a UV lamp, thus reducing the cost of detection as the other sources of light are not needed.

However, Honeychurch (2014) also warns against the possible “traps” which are caused by structural defects of QDs and which may not allow the electron-hole recombination

and thus preventing the occurrence of continuous fluorescence. The surface passivation in the form of the shell made from the other semiconducting material with wider band gaps is used in order to minimize the defects and enhance the photostability. Regardless the shell, it is always the size and the material of the core which determine the wavelength of emission.

4.2 QDs Synthesis

Several methods of QDs synthesis have been introduced, however, all of them seek for an optimal compromise between high photoluminescence and easy preparation together with the possibility of QDs to be used in biological application. Therefore, the synthesized QDs, in order to be used for biological detection, should be stable, water-soluble and, most importantly, non-toxic. These features can be adjusted during a synthesis process, which is, in general, divided into two approaches: bottom-up and top-down, while each approach includes several methods. As far as QDs are concerned, there are basically three methods of synthesis that are profoundly discussed by Drbohlavová (2009, 2015), namely the lithography-based method, epitaxial growth, and preparation of QDs via colloidal chemistry. Therefore, when describing the processes and their features, I will proceed from the above-mentioned researches.

As for the lithography-based methods, the synthesized QDs exhibit a poor quality of optical properties, therefore they are not suitable for optical detection. Moreover, these methods are considered as expensive and time-consuming processes.

Epitaxial growth techniques, on the other hand, offer various advantages over lithography-based methods, for example the possibility of controlling properties. Since epitaxial growth is “a process during which a crystal is formed on an underlying crystalline surface as the result of deposition of new material onto that surface” (Narlikar, 2010, p. 210), also the resulting position can be controlled. Using this technique, the self-assembling arrays of QDs in the shapes of islands are formed. Drbohlavová et al. (2009) highlight the application of such synthesized QDs mainly in the field of optoelectronics, however, she also mentions a possible future application of such QDs in biosensing. The main promising feature is seen in the possibility to arrange a sensor in that way that each QD would emit different wavelength, hence

enabling simultaneous detection of distinct analytes. Drbohlavová et al. (2009) report that colloidal QDs in the shape of a sphere are preferred in biological application, though.

Using colloidal chemistry, both hydrophilic and hydrophobic QDs can be synthesized, and the both methods involve the combination of metallic (for hydrophilic) or organometallic (for hydrophobic) precursor with chalcogen precursor as the essential elements. In both methods, the stabilization substance is added in order to prevent particle aggregation as well as to ensure stability even in high temperatures, because the corresponding solution is heated to a certain temperature for several hours, however, the degree of Celsius differs. It is also worth mentioning that the temperature during synthesis is one of the factors determining the resulting photoluminescence properties.

Drbohlavová (2015) further explains that during the synthesis of hydrophilic QDs the solution is heated to 100°C. The most prominent features of QDs synthesized with this method are water-solubility and low toxicity, but also a relatively inexpensive process is favourable, however, the intensity of photoluminescence is rather low with the quantum yield⁷ of 30%. On the contrary, in the case of the hydrophobic QDs, where the solution is heated up to 300°C, better photoluminescence with quantum yield of 60% is reported. Nevertheless, the insolubility in water would restrict the biological application, therefore further surface modifications are necessary.

4.2.1 Surface modification and functionalization

According to Drbohlavová et al. (2009), there are two main methods of hydrophilization, namely silanization and cap exchange, which also enable implementation of a functional group.

Silanization is a process of adding a shell of silicon oxide to the nanocrystal surface which enhances the stability of QDs, decreases toxicity and provides QDs with

⁷ The fluorescence quantum yield is the fraction of the number of emitted photons to the number of absorbed photons (Lakowitz, 1999).

bioconjugation sites, i.e. makes an attachment of a biomolecule to the quantum dot possible, as was also discussed by Drbohlavová (2015).

The second method, described by Honeychurch (2014) and Drbohlavová (2015), comprises of replacing a hydrophobic layer with ligand molecules. Such a molecule has one end reactive towards the QD surface, and the second end ensures water solubility and enables implementation of suitable functional groups. Such QDs, which are water-soluble and, moreover, enhanced with various functional groups which allows bioconjugation with proteins, antibodies, and enzymes, are suitable for detection in biological applications.

Furthermore, Honeychurch (2014) distinguishes two possible arrangements of conjugates depending on the relative size of QDs and biomolecules. In the first type, QDs is larger than the molecule and thus several molecules, even of different functionalities, can be attached to one quantum dot. On the contrary, the second type consists of a larger molecule that is surrounded by many small QDs resulting in facilitated detection of the given molecule caused by an intense fluorescent signal.

4.3 Application of QDs in sensor technology

As indicated in the previous chapters, QDs find, in particular, application in nanomedicine as a biosensor, however application in optoelectronics is of great interest as well. QDs as a nanosensor are above all used for optical detection mainly thanks to their favourable optical properties. Besides detection of toxic substance either in water or in living organisms, QDs are also used for detection of bacteria, viruses, and DNA strands, while in both cases the possibility of functionalizing the surface of QDs with miscellaneous biomolecules also plays an important role.

In those applications, usually colloidal QDs with bioconjugates, i.e. with an attached biomolecule, for instance protein or antibody, are preferred. Honeychurch (2014) also stresses that small QDs, usually of 2-4 nm in diameter, are used to conjugate with the above-mentioned biomolecules, because that way the biomolecules are not influenced by the marker and maintain their natural function in chemical processes

inside the organisms. Drbohlavová (2015) further explains that when a tissue with those QDs is illuminated, easy detection of movement and condition of biomolecules can be carried out. Moreover, if QDs of different size are used, the simultaneous distinction of diverse organelles, i.e. cell subunits, is possible on the basis of different emitted colour. Therefore, QDs are also extensively investigated and used as a tool for cell labelling and biological imaging.

As far as biosensors are concerned, Drbohlavová et al. (2009) emphasise the three types of biosensors which are of great interest: immunoassays, DNA and protein sensors, and sugar sensors. “Highly ordered QDs array may represent the optical (eventually electrochemical) transducer of miniaturized biosensor, which can be employed for *in vitro* detection of biogenous analytes via monitoring the change of QDs fluorescence properties” (Drbohlavová et al., 2014, p. 1602). In biological application, thus, both optical and electrochemical detection can be employed.

4.3.1 Optical detection

The already mentioned optical properties of QDs, such as photostability and narrow emission spectra, enable both real-time monitoring and simultaneous detection of various analytes, since the overlap of emitted colour is minimum. These advantages, as highlighted by Fojtík (2014), place QDs as a possible substitution of organic dyes, which do not exhibit such properties.

The application of QDs in immunoassays utilizes the conjugation of QDs with a specific antibody, which may be perspective as far as selective and sensitive detection is considered. In some cases, the simultaneous detection was achieved as well, for example Mihaljović (2014) mentions simultaneous detection of bacteria *Escherichia coli* and *Salmonella*, and Fojtík (2014) reports detection of breast cancer marker Her2, toxin of diphtheria, and toxin of tetanus at the same time.

One of the techniques extensively used for sensing in biological system is the FRET (Förster resonant energy transfer) method, which is defined by Bhushan (2012, p. 512) as “a resonant non-radiative energy transfer from a donor to an acceptor fluorophore due to the dipolar interaction.” In other words, when the donor (usually a fluorophore)

is irradiated, it emits energy in the form of light that is absorbed by the acceptor and thus stimulates the acceptor's fluorescence. However, the FRET occurs only if the donor-acceptor distance is within the range of 1 – 10 nm, and only if the emission wavelength of the donor overlaps with the absorption wavelength of the acceptor.

Many of optical properties of QDs, such as broad absorption spectra and narrow emission spectra which is also tuneable, makes them ideal as donors in the FRET system. Fojtík (2014) confirms that the employment of a QD as a donor results in amplifying the signal, thus lowering the detection limit. For instance, Bhushan (2012) mentions the application where a donor-acceptor couple of the FRET system is made from QDs and a quencher, respectively. A quencher, for instance, is a dye-labelled protein that suppresses QD emission. Upon binding the target analyte to the protein, the QD emission is recovered, as illustrated in Figure 7.

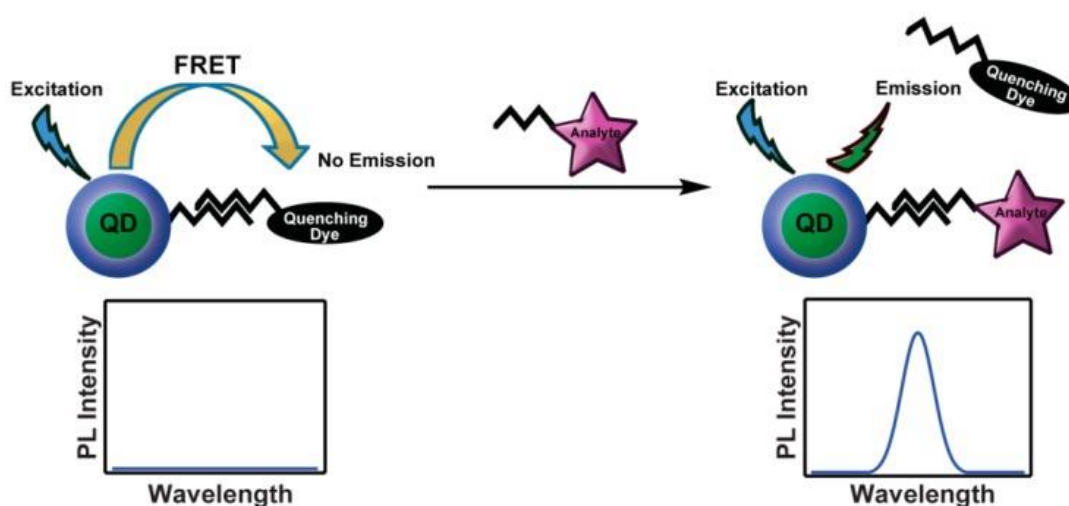


Figure 7. Recovery of QD emission upon target binding.

Reprinted from Shamirian (2015).

Bhushan (2012) evaluates this technique as a sensitive detection tool with high selectivity and also conveys the possibility to use QDs of different colour for simultaneous detection of various targets, such as bacteria, viruses, or to be used for DNA analysis. To give an example of such employment, Honeychurch (2014) introduces QDs (donor) and gold nanoparticle (acceptor, quencher) FRET pair for detection of *Staphylococcus aureus*. Also, the same FRET pair, functionalised with specific molecules, is reported by Honeychurch (2014) to be used as a glucose bioprobe

that enabling measure of glucose concentration on the basis of recovered QD's fluorescence.

4.3.2 Electrochemical detection

Fojtík (2014) describes this method for DNA hybridization monitoring, where the electrochemical dissolving analysis is applied. The analysis consists of two steps: an accumulation of target (e.g. QDs with an attached DNA) and voltammetric measuring of dissolved metal ions on the electrode. Fojtík (2014) stresses the property of Cd ions to be easily dissolved, therefore the CdS QDs markers amplify the dissolving signal and lower the detection limit.

4.3.3 Other QDs nanosensors

Besides application of QDs to nanobiosensors, there is a great deal of research which investigates the possibility of QDs to be used as temperature and pressure sensors, and as a fire detector sensors. Ke et al. (2016) explore low-pressure and temperature sensors based on QDs and their research show that in both cases the detection limit was decreased.

As for fire detectors based on QDs, De Iacovo (2017) conducted research where he employed colloidal QDs photoconductors for fire detection. His findings prove that QDs with their responsivity enable the detection of small flames even from a distance.

Those applications are still under development, though, which only supports the fact that scientists from all over the world are seeking for novel employment of nanostructures and their implementation into everyday life, although there are still many challenges to face.

5 Nanosensors: Advantages, opportunities, and challenges with a view to the future

Throughout this thesis, several advantages that distinguish nanotechnology-enabled sensors from the classical sensors have been mentioned, anyway, a summary list of them will be provided here. I will mainly consider three most appealing features that enhance sensor performance: sensitivity, selectivity, multiplexing.

The first one stems from the unique property peculiar to nanostructures which is a high surface-to-volume ratio. This property together with its enhancing effect on the reactivity of given material has been described in the first chapter of this thesis. Upon increased reactivity, the detection limit of transducer can be shifted to the detection of a single molecule, as it has been reported in some research mentioned in this thesis, e.g. Patolsky (2006). Such nanosensors will provide much more sensitive detection than the current sensors.

With the ability to bind a specific antibody to the surface of nanostructure, the selectivity of sensor is significantly increased enabling to detect specific target in the multi-element environment. In this thesis, I have mentioned the functionalization of QDs with various biomolecules which bind with the targeted analyte, and thus changing the optical properties of QDs.

The advantages of multiplexing, i.e. simultaneous detection of various targets, stems from the possibility to engineer nanostructures into arrays where each will be functionalized toward the different analyte. Such possible arrangement was mentioned when dealing with the application of nanowires, e.g. Patolsky's (2006) research, as well as with the application of QDs in immunoassays.

Besides mentioned advantages, or possibilities of nanotechnology-enabled sensors, other benefits, such as real-time detection, lower weight, and consequently less-power demand, are worth mentioning, since all of them makes nanosensors advantageous over the common sensors.

Although still under research and development, nanosensors with above listed advantages are expected to find their employment in various fields of industry and services. Evans (2009) mentions eight main sectors: medicine, national security, environmental monitoring, food industries, energy, transportation, workplace safety, and manufacturing. Despite all of them being equally possible application of nanosensors, I found particularly important the first four areas.

The opportunity of using nanosensors in medicine, which relates to biosensors, is quite profoundly described in this thesis. Biosensors, in general, benefit from nanoscale biosensors, since the interaction of molecules is on the nanoscale level. According to NanoMarket, LC (2014), the healthcare sector “is the largest initial market for nanosensors owing to an increasing requirement for rapid, compact, accurate and portable diagnostic sensing systems”. The fact that nanosensors are capable of fulfilling this necessity makes them a promising tool for the future preliminary diagnosis and detection of diseases markers. Fedel et al. (2016) introduce a nanosensing platform called Veridex’s CellSearch[®] based on magnetic nanoparticles that enable detection even of a small number of tumour cells in a small sample of blood, strictly speaking 5 cells in 7.5 ml of blood.

The second market sector, which is also mentioned in this thesis, is the national and military security, where early and precise detection of toxic or explosive gases, or airborne biological agent plays an important role. Current alarms and detectors do not expose such sensitivity, which may result in late detection. Wilson (2005) introduces a nano-enabled chemical sensor called SnifferSTAR[®] that can be integrated into a micro unmanned aerial vehicle used for early detection of chemicals without exposing anybody to danger.

Detection of chemicals is not only tool for the military security, but also a key factor for controlling air pollution. Moreover, besides air, nanosensors can find application in monitoring and providing detection of chemicals, like pesticides, organic dyes, or cleaning fluids, in water and land. NanoMarket, LC (2014) proposes another opportunity, which is utilization of nanosensors in food industry, mainly in monitoring the quality of food during the whole production process, from harvesting, packaging to marketing. Although it is still in its early stage, the main focus in this monitoring will

be placed on detection of pathogens which may cause food-borne illness, as it is further explained by Evans (2009).

All of the above-mentioned opportunities are very promising demonstrations how nanotechnology can be implemented into market and improving current technologies, however, there are some challenges that should be addressed.

As far as implementation to medicine and food industry is concerned, most importantly toxicity of nanoparticles should be considered. Because of the nanoscale, many nanostructures possess novel and beneficial properties, however, in many cases the long-term effects of such nanostructure on a human body are not studied profoundly. Fedel et al. (2016) address the need for standards that will help to provide normalized evaluation of sensor performance. The pre-testing and calibration should be a part of such standards used for defining nanosensors reliability, sensitivity, and other parameters.

Nevertheless, there is no doubt that the main challenge is the cost and low reproducibility of the nanomaterial's synthesis. The expensive price stems from the lack of manufacturing facilities, as well as from the lack of highly specialized workforce that would enable production of nanomaterial on a mass-scale at a reasonable price and within a short time. Wilson (2005) considers this fact as a major challenge that restricts implementation of nanosensors into commercial operation, however, Fedel et al. (2016) also appeal for a better communication and collaboration between the scientists and the industrial partners.

Although, there have been some commercially successful nanotechnology-enabled sensors, as mentioned above, the path from laboratory research to a commercially available product is still in the early stage. Yet, with advanced researches and with overcoming above-mentioned challenges, there is no doubt that nanosensors will play an important role in several market sectors.

Conclusion

The aim of this bachelor's thesis was to research and analyse available literature concerning nanostructures and their application as a sensing device, in particular, concerning the detection of biological and chemical analytes.

The thesis consists of five main chapters, however, with respect to its content, it can be roughly divided into three parts. The first part, covering the first and the second chapter, deals with the introduction to two fields of technology: nanotechnology and sensor technology, where the basic definition, principles, and parameters are described.

As for the second part (i.e. the third and the fourth chapter), the focus was placed on two specific nanostructures, a nanowire and a quantum dot, and covered the topics of fabrication and special properties of the given nanostructure. In the final part of the third and the fourth chapters, the particular application of a nanowire and of a quantum dot as a tool for sensor enhancement was explained, respectively.

Despite the short history of this technology, the progress made in this area is remarkable and nanosensors find application in many industries. Those opportunities, as well as challenges were discussed in the concluding part of this thesis, i.e. the fifth chapter.

Overall, this thesis provided the reader with the theoretical framework of both nanostructures and nanosensors, which can serve as an introductory survey for anybody interested in this modern technology.

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List of abbreviations

A/D converter – analog-to-digital converter

DNA – deoxyribonucleic acid

FET – field effect transistor

FRET – Förster resonant energy transfer

NW – nanowire

PNA – peptide nucleic acid

QD – quantum dot

RNA – ribonucleic acid

UV – ultraviolet

VLS method – vapor-liquid-solid method